

ARCO Chemical Company
Legal Department
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Newtown Square, Pennsylvania 19073-2387
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John C. Martin, Jr.
Senior Patent Counsel

November 21, 1995

John Kollar
President
Redox Technologies Inc.
6 Spencer Court
Wyckoff, NJ 07481

Re: Redox's EG Technology

Dear Mr. Kollar:

I acknowledge receipt of your October 31 letter relating to Redox's EG Technology.

Following our October 31 meeting, we reviewed earlier technical suggestions and enclose for your information a copy of Technical Suggestion 81-73 signed October 16, 1981 titled "Improved Process for the Production of Diteriary Butyl Peroxide." Preparation of DTBP is described by the reaction of either TBA or isobutylene using tertiary butyl hydroperoxide (TBHP) and an ion exchange acid resin catalyst. The first paragraph, second typed page of the Technical Suggestion states that "a large pore size resin would be preferable." As you know, large pore size resins are known as macroreticular resins.

Therefore, this Technical Suggestion meets the paragraph 6(b) exception to confidentiality of the April 23, 1987 agreement in that the information disclosed to ARCO Chemical Company was in its possession prior to receipt from Redox.

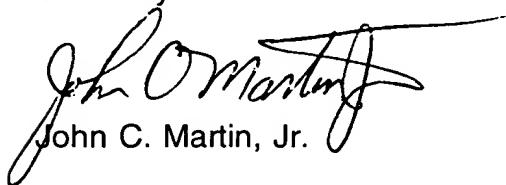
As I stated at our October 31 meeting, we are willing, at your request, to provide affidavits from all named inventors of our 5,371,298 patent stating that they did not have access to any of Redox's technology (i.e., their invention was independently developed from your information). See paragraph 6(d) of the April 23, 1987 agreement.

John Kollar
November 21, 1995
Page 2

Some of us believe that using macroreticular resins for this reaction is in the publicly available literature such as early Rohm & Haas Company product bulletins describing uses of macroreticular resins. To date, such literature has not been uncovered.

In any event, the information already provided to you by this letter and at our October 31 meeting clearly eliminates any possible claim by Redox against ARCO Chemical Company under the April 23, 1987 agreement.

Sincerely.



John C. Martin, Jr.

cc: W. J. Klingebiel

Enclosure

T. S. No. 81/13

Date Assigned

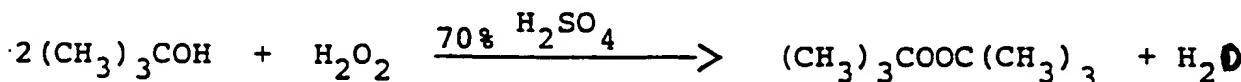
10/16/81

USE PENCIL OR TYPEWRITER - Suggesting form for disclosure: (1) Title of invention, (2) State the precursors prior to your invention, (3) Problems and difficulties in connection with those prior practices, and how your invention solves these problems, (4) Your solution or invention - describe fully in print, with drawings, diagrams, and structural formulas, whom you will disclose, (5) Give test results illustrating merits of your invention, (6) List reference pages of laboratory notebook and the persons to whom the invention has been disclosed, (7) Names of inventors, and (8) Cross out unused space. (9) Forward disclosure to Patent Department.

(1) Title of Invention Improved Process for the Production of Diteriary Butyl Peroxide

(2) - (3) - (4) - (5)

Diteriary butyl peroxide (DTBP) is a specialty product used primarily as an organic peroxide catalyst or catalyst intermediate. It is commercially produced by the reaction of pure t-butanol (TBA) or isobutylene with hydrogen peroxide in the presence of concentrated sulfuric acid:



The process requires refrigeration to control the reaction isotherm ($\Delta H = 98,000$ Btu/lb mole) at or below 15°C thus reducing side reactions and TBA dehydration. The product mixture is quenched with excess water to promote phase separation. The organic phase, consisting of mostly DTBP, is washed with dilute alkali solution and the aqueous acid phase is usually neutralized and discarded.

When excess hydrogen peroxide is used in the above reaction, a mixture tert-butyl hydroperoxide and DTBP is obtained.

An alternative process, that has been experimentally demonstrated and recently evaluated (1), substitutes tert-butyl hydroperoxide for the hydrogen peroxide:

Project No. _____

(6) Test data and original references may be found on the following notebook pages 187434, 187435.

Inventor(s) Signature

E. A. Hazbun *E. A. Hazbun*

Date signed

10/13/81

Date of Conception

Date signed

Date of Conception

Signature of Witness

V. M. Chong

Date signed

10/15/81

Date first disclosed to me

Signature of Witness

J. A. Cahill

Date signed

10/15/81

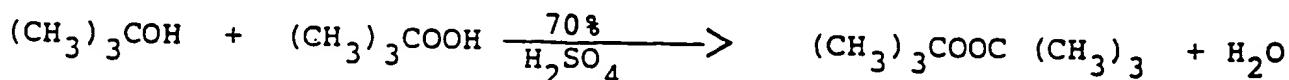
Date first disclosed to me

T. S. No. 6

Date Assigned 10/16/81

(1) Title of Invention Improved Process for the Production of Ditertiary Butyl Peroxide

$$(2) + (3) + (4) = (5)$$



This also requires refrigeration and acid neutralization. The estimated transfer price via this process at the 3 MM lbs/yr scale is \$1.16/lb for pure DTBP.

Recent preliminary testing of DTBP as a cetane improver for diesel fuels (2) have shown the following encouraging results:

V %	Cetane #
0	44.8
3	56.8
5	67.8

Such a performance may create a large volume application for DTBP, if it can be manufactured at low cost.

To allow continuous production at low cost, I propose that DTBP be produced by the reaction of TBHP and TBA over an ion exchange acid resin catalyst at a temperature sufficiently elevated to permit cooling with cooling tower water obviating the need for refrigeration.

(1) Title of Invention Improved Process for the Production of Ditertiary Butyl Peroxide

(2) - (3) - (4) - (5)

$$(\text{CH}_3)_3\text{COH} + (\text{CH}_3)_3\text{COOH} \xrightarrow[\text{H}_2\text{SO}_4]{70\%} (\text{CH}_3)_3\text{COOC}(\text{CH}_3)_3 + \text{H}_2\text{O}$$

This also requires refrigeration and acid neutralization. The estimated transfer price via this process at the 3 MM lbs/yr scale is \$1.16/lb for pure DTBP.

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To allow continuous production at low cost, I propose that DTBP be produced by the reaction of TBHP and TBA over an ion exchange acid resin catalyst at a temperature sufficiently elevated to permit cooling with cooling tower water obviating the need for refrigeration.

Project No. _____	(6) Test data and original references may be found on the following notebook pages <u>187435</u> <u>187436</u>		
Inventor(s) Signature		Date signed 10/13/81	Date of Conception -----
Signature of Witness		Date signed 10/15/81	Date first disclosed to me -----
Signature of Witness		Date signed 10/15/81	Date first disclosed to me -----
Signature of Witness		Date signed 10/15/81	Date first disclosed to me -----

Page 2 of 4 pages

M P O N T A N T

(1) Title of Invention Improved Process for the Production of Ditertiary Butyl Peroxide

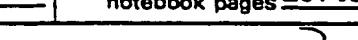
(2) + (3) + (4) + (5)

Based on past experience with acid resin catalysts for MTBE and SBA production, these catalysts tend to yield more moderate reaction rates at higher selectivity than concentrated sulfuric or other mineral acid. A large pore size resin would be preferable in this instance.

The reactor may be a multistage packed bed with interstage cooling or a multitube heat exchange design.

Since the purity requirements for a diesel fuel additive may not be stringent, Isobutane oxide, consisting of near equimolar proportions of crude TBA and TBHP, may be used as reaction feed thus lowering raw materials cost.

The reaction is driven to completion and the resulting crude DTBP - water mixture may be washed with water or slightly alkaline solution to remove soluble formates, aldehydes and ketones originally present in the feed; trace acid from the resin catalyst is also washed or neutralized in this step. The DTBP is then phase separated and dried (if required). Because the reactor effluent does not contain sulfuric acid, the caustic quench and neutralization is eliminated.

Project No. _____	(6) Test data and original references may be found on the following notebook pages <u>187436</u> , <u>187437</u> , _____		
Inventor (s) Signature <u>E. A. Hazbun</u> 	Date signed <u>10/13/81</u>	Date of Conception	
	Date signed	Date of Conception	
Signature of Witness <u>V. M. Chong</u>	Date signed <u>10/15/81</u>	Date first disclosed to me	
Signature of Witness <u>J. A. Cahill</u>	Date signed <u>10/15/81</u>	Date first disclosed to me	

Chemical Company 

Technical Suggestions

T. S. No. 81-7 Date Assigned 10/16/81

Improved Process for the Production of Ditertiary Butyl Peroxide

(2) + (3) + (4) + (5)

Such a process could yield DTBP of moderate purity at a transfer price in the 30 to 50¢/lb range depending on plant scale.

References

1. DTBP Production, R. L. Bobeck to B. Cramer, Aug. 1, 1979.
2. Initial Testing of DTBP as Octane Improver, J. M. DeJovine to J. A. Tarengelo, Jan. 14, 1981. Also U. S. Patent 2,378,341.

USE PEN OR TYPEWRITER - Suggested form for disclosure: (1) Title of Invention, (2) State the practices prior to your Invention, the problems in connection with these prior practices, and how your Invention solves these problems, (3) Your solution or Invention - describe fully in prose, with diagrams and structural formulas, when appropriate, (4) Give test results illustrating merits of your Invention, (5) Describe principal advantages over prior practices, realizing by your Invention, (6) List reference pages of laboratory notebook and the persons to whom the Invention has been disclosed, (7) Names of inventors (witnesses are recorded on each sheet of the disclosure), (8) Cross out unused space, (9) Forward disclosure to Patent Department.

IMPOR TANT

Project No. _____	(6) Test data and original references may be found on the following notebook pages <u>187437</u>		
Inventor(s) Signature	<p>E. A. Hazbun <u>E. A. Hazbun</u></p> <p>Date signed <u>10/13/81</u></p> <p>Date of Conception _____</p>		
Signature of Witness	<p>V. M. Chong <u>V. M. Chong</u></p> <p>Date signed <u>10/15/81</u></p> <p>Date first disclosed to me _____</p>		
Signature of Witness	<p>J. A. Cahill <u>J. A. Cahill</u></p> <p>Date signed <u>10/15/81</u></p> <p>Date first disclosed to me _____</p>		

OBJECT OF WORK

Improved process for the production of tertiary butyl peroxide.1.5 81-13 10/16/81 work

DATE

Olelkert C. J. & Cahill - October 16, 1981

Tertiary butyl peroxide (DTBP) is a specialty product used primarily as an organic peroxide catalyst or catalyst intermediate. It is commercially produced by the reaction of pure t-butanol (TBA) or isobutylene with hydrogen peroxide in the presence of concentrated sulfuric acid:



The process requires refrigeration to control the reaction isotherm ($\Delta H = 98,000 \text{ Btu/lb-mole}$) at or below 15°C thus reducing side reactions and TBA dehydration. The product mixture is quenched with excess water to promote phase separation. The organic phase, consisting of mostly DTBP, is washed with dilute alkali solution and the aqueous acid phase is usually neutralized and discarded.

When excess hydrogen peroxide is used in the above reaction, a mixture tert-butyl ^{hydro}peroxide and DTBP ~~is~~ obtained.

SIGNATURE

E. F. Harlan

DATE 10/13/81

WORK CONTINUED ON PAGE 187435

WITNESSED AND UNDERSTOOD BY

Victor M. Chory
J. A. Cahill

DATE 10/15/81

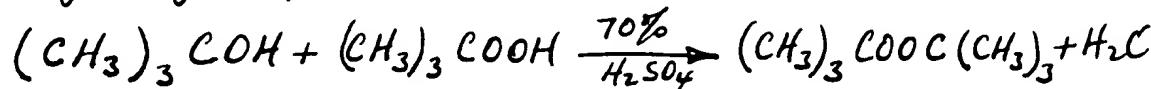
10/15/81

OBJECT OF WORK Improved process for the production of
dibutyl peroxide

TS 81-73 10/16/81 Cont'd.

DATE

An alternative process, that has been experimentally demonstrated and recently evaluated (1), substitutes tert-butyl hydroperoxide for the hydrogen peroxide:



This also requires refrigeration and acid neutralization. The estimated transfer price via this process at the 3 MM lbs/yr scale is \$ 1.16/lb ~~for~~ pure DTBP.

Recent preliminary testing of DTBP as a cetane improver for diesel fuels (2) have shown the following encouraging results:

✓ %	Cetane #
0	44.8
3	56.8
5	67.8

Such a performance may create a large volume application for DTBP if it can be manufactured at low cost.

SIGNATURE

E. F. Hargrave

DATE 10/13/81

WORK CONTINUED ON PAGE 187434

WITNESSED AND UNDERSTOOD BY

*Victor McHenry**J. A. Cooley*

DATE

10/15/81

10/15/81

OBJECT OF WORK

Improved process for the production of
diteriary butyl peroxide.

T.S. 81-73 10/16/81 cont'd.

DATE

J. A. Calfee - October 16, 1981

To allow continuous production at low cost, I propose that DTBP be produced by the reaction of TBHP and TBA over an ion exchange acid resin catalyst at a temperature sufficiently elevated to permit cooling with cooling tower water obviating the need for refrigeration.

Based on past experience with acid resin catalysts for MTBE and SBA production, these catalysts tend to yield more moderate reaction rates at higher selectivity than concentrated sulfuric or other mineral acid. A large pore size resin would be preferable in this instance.

The reactor may be a multistage & packed bed with interstage cooling or a multi-tube heat exchange design.

Since the purity requirements for a diesel fuel additive may not be stringent, Isobutane oxidate, consisting of ~~near~~ equimolar

SIGNATURE

E. F. Hartman

DATE 10/14/81

WORK CONTINUED ON PAGE 187437

WITNESSED AND UNDERSTOOD BY

Victor M. Chong

DATE

10/15/81

J. A. Calfee

10/15/81

OBJECT OF WORK Improved process for the production of
diteriary butyl peroxide

T.S. 81-73 10/16/81 Excluded

DATE	October 16, 1981 - Fletcher E. Myrick
	<p>proportions of crude TBA and TBHP, may be used as reaction feed thus lowering raw materials cost.</p> <p>The reaction is driven to completion and the resulting crude DTBP - water mixture may be washed ^{with water or} in slightly alkaline solution to remove soluble formates, aldehydes and ketones originally present in the feed; trace acid from the resin catalyst is also washed or neutralized in this step. The DTBP is then phase separated and dried (if required). Since Because the reactor effluent does not contain sulfuric acid, the ^{caustic} quench and neutralization is eliminated.</p> <p>Such a process could yield DTBP of moderate purity at a transfer price based in the 30 to 50 \$/lb range depending on plant scale.</p> <p><u>References</u></p> <ol style="list-style-type: none">1- DTBP Production, R.L. Bobeck to B. Cramer, Aug 1st, 19792- Initial testing of DTBP as Octane Improver J.M. DeJouine to J.A. Tarengelos Jan 14, 1981. Also US Patent 2,378,341

SIGNATURE E. F. Hazlwood DATE 10/14/81 WORK CONTINUED ON PAGE
WITNESSED AND UNDERSTOOD BY Victor McHenry DATE 10/15/81
J. A. Colwell DATE 10/15/81